

Quantum Condensation from a Tailored Exciton Population in a Microcavity

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An experiment is proposed, on the coherent quantum dynamics of a semiconductor microcavity containing quantum dots. Modeling the experiment using a generalized Dicke model, we show that a tailored excitation pulse can create an energy-dependent population of excitons, which subsequently evolves to a quantum condensate of excitons and photons. The population is created by a generalization of adiabatic rapid passage, and then condenses due to a dynamical analog of the BCS instability.

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There is great interest in the possibility of quantum-condensed phases of solid-state quasiparticles, such as excitons, polaritons, and magnons. Such phases are characterized by the presence of a quantum state whose population scales with the size of the system, and hence is much larger than one – macroscopic occupation. This is seen in recent experiments on Bose-Einstein condensation (BEC) of polaritons and polariton lasing. In these experiments [1, 2, 3, 4, 5], a semiconductor microcavity is excited at high energies, and a macroscopic population of low-energy polaritons emerges following relaxation and inelastic scattering [6]. These condensates appear spontaneously, from states without macroscopic occupations. This differentiates them from the microcavity parametric oscillator experiments [7], where resonant pumping of polaritons leads directly to a macroscopic occupation.

The aim of this paper is to show how microcavities could be used to access condensation, even in the absence of relaxation or inelastic scattering. We propose an experiment on a microcavity containing an ensemble of quantum dots, where the exciton decay times are many tens or hundreds of picoseconds [8]. We demonstrate that this experiment could be faster than these decay times, so that energy relaxation and inelastic scattering would be negligible. Nonetheless, we shall show that a condensate develops. In contrast with a laser, this condensate is formed from part-matter, part-light quasiparticles. In contrast with the microcavity parametric oscillator, it develops from a state with no macroscopic occupations, in the absence of the pump laser. And whereas relaxation is essential to obtain an equilibrium BEC or polariton laser, in our approach condensation occurs due to an instability of the coherent quantum dynamics. Our proposal implements, in a solid-state system, the type of dynamical condensation predicted in quenched Fermi gases [9, 10, 11, 12].

The first stage in our proposed experiment is the creation of a population of excitons in the quantum dots. We propose using a chirped laser pulse, which sweeps up through part of the inhomogeneously-broadened exciton line. As shown by the demonstration of Rabi oscilla-

tions [13] and density-matrix tomography [14], excitons in quantum dots are discrete two-level systems, which can therefore be manipulated using laser pulses. The proposed pulse implements adiabatic rapid passage, which is a well-established technique for populating discrete states [15]. It extends the technique, by controlling the pump spectrum to create an energy-dependent exciton population (Fig. 2).

The second stage occurs after the pump pulse has passed. It is the coherent quantum dynamics of the system, starting from the exciton population created by the pump. The pump is chosen such that this population is similar to a Fermi distribution, with a sharp upper step. The system is described by a model similar to that which describes pair condensation in atomic gases and superconductors. We therefore expect that a population with the form of a Fermi distribution could condense, due to a dynamical version of the BCS instability [12].

We now turn to the theoretical demonstration of this proposal. For simplicity, we suppose that the pump is circularly polarized, so we may consider only one of the polarization states of the excitons. We model the quantum dots as a set of two-level systems, each describing the presence or absence of an exciton of the pump polarization in a given localized dot state. These localized excitons are coupled to the electromagnetic field by the dipole interaction. Since the exciton states in the dots are spatially separated, we neglect the non-radiative interactions between the different two-level systems. The appropriate Hamiltonian is then the generalized Dicke model [16].

We label the dot states with an index i , so that E_i is the energy of an exciton in the i^{th} dot state; this state is localized at \mathbf{r}_i , with dipole-coupling strength g_i . The area density of dots is n , so that if $g_i = g$ and $E_i = E$ the vacuum Rabi splitting at resonance is $2g\sqrt{n}$. The state of a dot is specified by the Bloch vector $\langle\sigma\rangle$, where $\sigma_i^- = \sigma^x - i\sigma^y$ is the exciton annihilation operator, and σ^z the inversion. The inversion is related to the occupation of the dot n_i by $n_i = (\sigma^z + 1)/2$, and is $-1(+1)$ for an unoccupied (occupied) dot.

Dicke models have previously been used to describe polariton condensation in equilibrium [2, 16, 17], and in a dissipative open system [18]. Here we are concerned with the opposite limit, of timescales short compared with the relaxation times. The dynamics therefore obeys the Heisenberg equation. Since we are concerned with condensation phenomena, involving large photon numbers, we treat the field classically. However, we retain the full quantum dynamics of the dots, and hence the possibility of an incoherent population of excitons. In this approximation, the Heisenberg equation gives

$$i\dot{\psi}_{\mathbf{k},t} = \omega_{\mathbf{k}}\psi_{\mathbf{k},t} + \frac{1}{2} \int g P_{\mathbf{k},t}(E, g) dE dg + F_{\mathbf{k},t}, \quad (1a)$$

$$i\dot{P}_{\mathbf{k},t}(E, g) = EP_{\mathbf{k},t}(E, g) - 2g \sum_{\mathbf{k}'} D_{\mathbf{k}-\mathbf{k}',t}(E, g) \psi_{\mathbf{k}',t}, \quad (1b)$$

$$i\dot{D}_{\mathbf{k},t}(E, g) = g \sum_{\mathbf{k}'} (\psi_{\mathbf{k}',t}^* P_{\mathbf{k}+\mathbf{k}',t}(E, g) + P_{\mathbf{k}'-\mathbf{k},t}^*(E, g) \psi_{\mathbf{k}',t}). \quad (1c)$$

$\psi_{\mathbf{k}}$ is the normal-mode amplitude for an electromagnetic field mode, with in-plane wavevector \mathbf{k} and energy $\omega_{\mathbf{k}}$, and $F_{\mathbf{k}}(t)$ is the (classical) pump. $P_{\mathbf{k},t}(E, g)$ and $D_{\mathbf{k},t}(E, g)$ are collective variables describing the polarizations and inversions of the dots. They are defined as

$$P_{\mathbf{k}}(E, g) \delta E \delta g = \frac{1}{An} \sum_i' \langle \sigma_i^- \rangle e^{-i\mathbf{k} \cdot \mathbf{r}_i}, \quad (2)$$

$$D_{\mathbf{k}}(E, g) \delta E \delta g = \frac{1}{An} \sum_i' \langle \sigma_i^z \rangle e^{-i\mathbf{k} \cdot \mathbf{r}_i}, \quad (3)$$

where the sums run over the states with $E \rightarrow E + \delta E$ and $g \rightarrow g + \delta g$.

Eqs. (1a–1c) generalize the Maxwell-Bloch equations [19], to allow for the distribution of energies and dipole-coupling strengths in the dots. Thus the inversion and polarization become distribution functions: $D_0(E, g) \delta E \delta g$ is the inversion due to states with energies $E \rightarrow E + \delta E$ and couplings $g \rightarrow g + \delta g$.

The fields $P_{\mathbf{k},t}$, $\psi_{\mathbf{k},t}$, and $F_{\mathbf{k},t}$ have been normalized such that their square magnitudes are particle numbers per exciton state. A condensate is characterized by a macroscopic occupation number, *i.e.*, one which scales with the size of the system. Thus an exciton-photon condensate has at least one $P_{\mathbf{k},t} \sim N^0$ and one $\psi_{\mathbf{k},t} \sim N^0$, where N is the total number of dots in the active region of the sample. In contrast, in a non-condensed state there is at most of order one particle per mode, and the $P_{\mathbf{k},t}$ and $\psi_{\mathbf{k},t}$ are all $\lesssim N^{-1/2}$.

The approximation leading to (1) is the standard semiclassical approximation, used to treat condensates including lasers [20], superconductors, BCS superfluids [9, 10], and polariton condensates [21]. It neglects the

quantum fluctuations of the electromagnetic field, which dominate if the photon number is small, *i.e.*, close to threshold with a small number of dots [20, 21]. Since we could have $N \sim 10^3$, the semiclassical approximation is in general very well-controlled. However, to obtain a correct description of the dynamics, we must supplement (1) with noise terms. Without such terms, the non-condensed solution remains a steady-state above threshold, as in all semiclassical treatments of condensation. However, it becomes unstable, and hence is not realized. We therefore add a perturbation driving the field into (1a), modeling for example spontaneous emission into the cavity modes. The form and strength of this perturbation does not affect our results: we show results with Gaussian white noise, but have obtained similar results using a delta-like kick.

We now specialize (1) to develop a simulation of our proposed experiment. We introduce imaginary parts to $\omega_{\mathbf{k}}$ to allow for the decay of the microcavity photons, with timescales of a few picoseconds [1]. The initial condition is $\langle \sigma_i^z \rangle = -1$, and there are many exciton states distributed over the active area of the sample. Thus the sum in (3) is strongly peaked near $\mathbf{k} = 0$, and we approximate the initial conditions as $D_{\mathbf{k},t}(E, g) = \delta_{\mathbf{k}} D_{0,t}(E, g)$. The dynamics is then that of a continuous medium due to motional narrowing, with the short-range spatial structure of the exciton states averaged out on the long scales of the photons.

We consider a plane-wave pump, at a high angle where the excitons lie outside the stop-bands of the mirrors. The field acting on the dots at this wavevector, \mathbf{k}_p , may then be taken as the driving field. Anticipating our analysis of the condensation, we retain only one other mode of the field, specifically the confined cavity mode with $\mathbf{k} = 0$. This reduces (1) to

$$i\dot{\psi}_0 = \omega_0 \psi_0 + \frac{1}{2} \int g P_0(E, g) dE dg, \quad (4a)$$

$$i\dot{P}_0(E, g) = EP_0(E, g) - 2g D_0(E, g) \psi_0, \quad (4b)$$

$$i\dot{P}_p(E, g) = EP_p(E, g) - 2g D_0(E, g) F_p, \quad (4c)$$

$$i\dot{D}_0(E, g) = g (F_p^* P_p(E, g) + P_p^*(E, g) F_p + \psi_0^* P_0(E, g) + P_0^*(E, g) \psi_0), \quad (4d)$$

corresponding to an ensemble of two-level systems, interacting with two modes of the field. P_p is the polarization at the pump wavevector, and F_p the driving field.

We have simulated our proposed experiment by solving (4) numerically, with $N = 4500$ two-level systems. Results are shown in Figs. 1–3. These results focus on a model with a single coupling strength g , and a Gaussian distribution of exciton energies with variance σ^2 . The pump is a linearly-chirped Gaussian,

$$2gF_p(t) = \frac{S}{\sqrt{2\pi\tau^2}} e^{-i(\nu_0 + \alpha t/2)t} e^{-t/(2\tau^2)}, \quad (5)$$

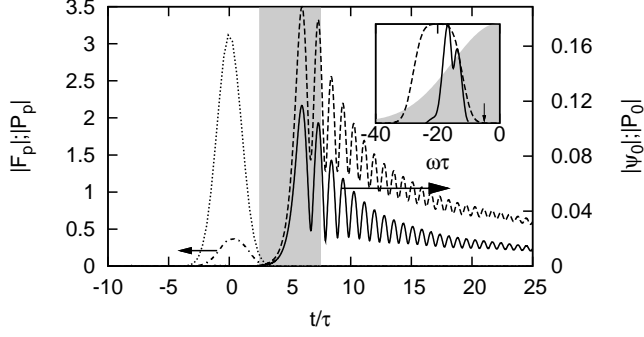


FIG. 1: Electromagnetic fields and polarizations as functions of time, for the simulation described in the text. Dotted: pump field $|F_p|$ (left axis). Dot-dashed: polarization $|P_p|$ at pump wavevector, integrated over dot energies and couplings (left axis). Solid: cavity field $|\psi_0|$ at $\mathbf{k} = 0$ (right axis). Dashed: polarization $|P_0|$ at $\mathbf{k} = 0$ (right axis). Inset: spectrum $|\psi_0(\omega)|^2$ during the shaded region of the main plot (solid curve). The pumped population (dashed curve), exciton energy distribution (shading), and energy of the $\mathbf{k} = 0$ cavity mode (arrow) are shown for comparison.

where $S = \int 2g|\psi(t)|dt$ is the usual pulse area per exciton state. The pulse time τ defines dimensionless times and energies, and the zero of energy is chosen at the center of the exciton line. The remaining parameters are $\sigma = 15\hbar/\tau$, $\Re(\omega_0) = -5\hbar/\tau$, $\nu_0 = -20\hbar/\tau$, $\alpha = 5/\tau^2$, $-\Im(\omega_0) = 1.5/\tau$, $g = 13/\tau$ and $S = 5\pi$. These parameters, with $\tau = 3$ ps, are reasonable for a microcavity containing interfacial quantum dots. Though there is a distribution of g , due to the different sizes of the dot states, this does not qualitatively change our results.

Figs. 1–3 are the key results of this paper, demonstrating the scenario outlined in our introduction. Referring first to Fig. 1, we see that there can indeed be two stages to the dynamics. In the first stage, during the pump pulse, P_0 and ψ_0 are vanishingly small. P_p does become finite, reflecting the fact that the pump laser does induce some coherent polarization in the excitons. This coherence is small – $P_p \approx 1/\sqrt{N} \approx 0.02$ – but more importantly both P_p and F_p have disappeared by the end of the pumping stage. We therefore argue that the pump produces an incoherent population of excitons. Furthermore, as shown in Fig. 2, this population has a sharp upper step, like that of the Fermi function at a low effective temperature $T \sim 1/\tau$.

In the second stage, visible in Fig. 1, we see both ψ_0 and P_0 building up to values of order N^0 . This directly demonstrates condensation of excitons and photons.

To understand the condensation, we consider the dynamics of the pumped population. Linearizing (1) gives normal modes $\psi_{\mathbf{k},t} = e^{i\lambda_{\mathbf{k},t}} A_\psi$, $P_{\mathbf{k},t} = e^{i\lambda_{\mathbf{k},t}} A_P(E, g)$. Their frequencies $\lambda_{\mathbf{k}} = \lambda'_{\mathbf{k}} + i\lambda''_{\mathbf{k}}$ obey [12]

$$\omega_{\mathbf{k}} - \lambda_{\mathbf{k}} = - \int \frac{\nu(E)}{E - \lambda_{\mathbf{k}}} dE, \quad (6)$$

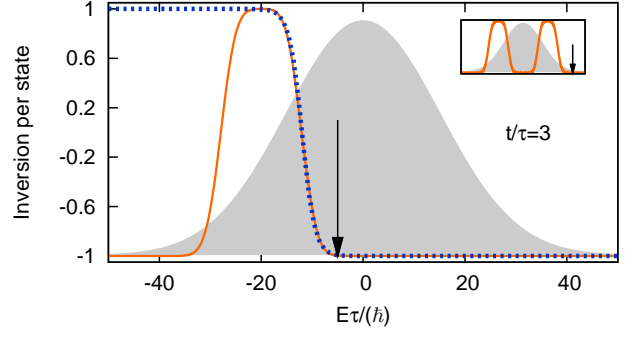


FIG. 2: (Color online) Simulated exciton inversion profile immediately after pumping, showing the population created by the Gaussian pump pulse (5) (solid line), and by a superposition of such pulses (inset). The dotted curve is an equilibrium exciton distribution with fitted temperature $\hbar/(4.2k\tau)$; this is 0.6 K for $\tau = 3$ ps. Arrows mark the energy of the $\mathbf{k} = 0$ cavity mode.

where $\nu(E) = \int g^2 D_0(E, g) dg$ is an optical density of the excitons. Applying this result to the state immediately after the pump pulse, we find an unstable mode at $\mathbf{k} = 0$. This instability gives the exponential growth of the polarization and field, to macroscopic values, visible in Fig. 1. In fact, (6) predicts instabilities for $|\mathbf{k}| < k_c$. The fastest-growing instability is at $\mathbf{k}=0$, and as this mode grows, it suppresses the gain for the others. It will thus be dynamically selected, and we therefore neglected cavity modes with $\mathbf{k} \neq 0$ in the simulations.

For the parameters used here, the instability predicted by (6) corresponds to the BCS instability in a superconductor. This can be seen by considering (6) for a single coupling strength, close to an instability. The eigenenergies $\lambda'_{\mathbf{k}}$ and growth rates $\lambda''_{\mathbf{k}}$ then obey

$$\frac{\omega'_{\mathbf{k}} - \lambda'_{\mathbf{k}}}{g^2} = -\mathcal{P} \int \frac{D_0(E)}{E - \lambda'_{\mathbf{k}}} dE, \quad (7a)$$

$$\lambda''_{\mathbf{k}} = \pi g^2 \text{sgn}(\lambda''_{\mathbf{k}}) D_0(\lambda'_{\mathbf{k}}) - \gamma. \quad (7b)$$

(7b) describes the growth or decay of the normal mode, with the first term the gain/loss from the excitons, and the second the loss due to the cavity decay $\gamma = -\Im(\omega_{\mathbf{k}})$. (7a) is the Cooper equation of the BCS model [22]. The term corresponding to the usual pairing interaction is $g^2/(\lambda_{\mathbf{k}} - \omega_{\mathbf{k}})$, which we recognize as the effective interaction between excitons, mediated by the cavity modes. Here it is an attractive interaction, as required for BCS, since the excitons lie below the photons. The term corresponding to the Fermi distribution is the exciton population created by the pump. In a superconductor, there is a solution to the Cooper equation below the Fermi energy, due to the step in the Fermi distribution; in the same way, (7a) has a solution below the step in the exciton occupation. (7b) shows that this mode experiences gain, and hence can become unstable.

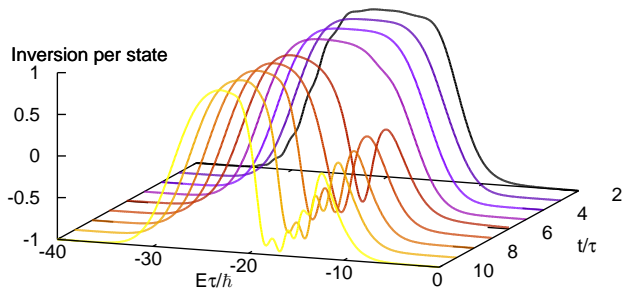


FIG. 3: (Color online) Simulated exciton inversion profile during the condensation shown in Fig. 1.

To confirm the origin of the condensation, we show in the inset to Fig. 1 the spectrum of the $\mathbf{k} = 0$ field, and in Fig. 3 the evolution of the population during condensation. As expected, the condensate is at a frequency below the step in the exciton population (Fig. 1); this leads to hole-burning there (Fig. 3). Reducing the center frequency of the chirp, ν_0 , we find that the condensation stage in Fig. 1 disappears, as the effective pairing interaction decreases below its critical value. The long-time limit of the simulations is then just the population of excitons. We have also confirmed that the phase of the pump (5) is irrelevant, by plotting the phase associated with the power spectrum in the inset to Fig. 1. This plot is identical in simulations with different pump phases, but the same random noise.

Since our condensate occurs on timescales short compared with the relaxation times, it will not be in equilibrium. This leads to phenomena absent for an equilibrium condensate. In Fig. 1, for example, we see ringing oscillations (corresponding to those predicted for atomic gases [9, 10, 11, 12]; unrelated oscillations have been predicted in coherently-driven microcavities [23]), and a slow decay. These phenomena will be discussed in a future publication.

It is perhaps surprising that our pump configuration could generate an incoherent population, without macroscopic occupation. To explain the mechanism, we note first that the driving field dominates during the pump stage (Fig. 1). Thus the pumping can be understood in terms of the well-known dynamics of non-interacting two-level systems, driven by a chirped laser pulse [15]. After eliminating the time-dependent frequency $\omega(t)$ of the pump with a unitary transformation, this dynamics is a precession of the Bloch vectors $\langle \sigma_i \rangle$ around axes $\mathbf{B}_i = (2g_i R(t), 0, \omega(t) - E_i)$, where $R(t)$ is the pump field at the dot.

Consider first a dot with energy inside the chirp. Provided the pump is strong enough, the Bloch vector of such a dot adiabatically follows \mathbf{B}_i from $-\hat{\mathbf{k}}$ to $\hat{\mathbf{k}}$, and it becomes populated. Furthermore, a dot with energy

outside the range of the chirp will not respond unless the pump is very strong, and hence such a dot will remain unpopulated. Thus we see that the only dots which could be polarized by the pump are those at the edge of the chirp. However, there are very few such dots. Furthermore, any polarizations induced in dots with different energies would have different phases, due to the chirping. Thus, as shown in Fig. 1, the chirped pump does not induce a collective polarization. Noting additionally that there are no cavity modes resonant with the pump, we see why the pump generates a state with $P_{\mathbf{k},t}, \psi_{\mathbf{k},t} \lesssim N^{-1/2}$.

A fully quantitative description of our proposed experiment is likely to require a more detailed model of the quantum dot states. It would also be desirable to extend our model to (a) incorporate relaxation processes, which on longer timescales will drive the off-equilibrium condensate towards an equilibrium one [12], and (b) develop a full quantum theory, as has been done for photon lasers [20], polariton lasers [5, 24], and equilibrium condensates [21]. Finally, it would be interesting to compare this work with Ref. [25], which appears to show a type of dynamical condensation, under very different conditions.

Though the parameters used here have been chosen to suit interfacial quantum dots [8, 26], many other choices are possible. The basic requirements are that the pumping is slow enough to create a controlled inversion profile within the inhomogeneous line, such that the couplings can be increased so that the instability occurs after the pump pulse, but before the excitons decay. Other systems described by generalized Dicke models, such as Fermi gases, SK dots in microcavities, or Josephson junction arrays, could be considered.

To conclude, we have proposed and analyzed a new approach to quantum condensation in a solid-state system. The key is the chirped pumping (5), which we have shown can create an energy-dependent population in the exciton line. Such a population can condense, even in the absence of relaxation or inelastic scattering. Since our approach uses the spectrum of the pump to tailor the population, it would be possible to pump other initial states (inset to Fig. 2). Thus our technique could be used more generally, to explore the quantum dynamics of a many-particle system from controlled initial conditions.

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- [1] H. Deng, D. Press, S. Gotzinger, G. S. Solomon, R. Hey, K. H. Ploog, and Y. Yamamoto, *Phys. Rev. Lett.* **97**, 146402 (2006).
 - [2] J. Kasprzak, M. Richard, S. Kundermann, A. Baas, P. Jeambrun, J. Keeling, F. Marchetti, M. Szymanska, R. Andre, J. Staehli, et al., *Nature* **443**, 409 (2006).
 - [3] R. Balili, V. Hartwell, D. Snoke, L. Pfeiffer, and K. West, *Science* **316**, 1007 (2007).

- [4] D. Bajoni, P. Senellart, E. Wertz, I. Sagnes, A. Miard, A. Lemaître, and J. Bloch, *Phys. Rev. Lett.* **100**, 047401 (2008).
- [5] A. P. D. Love, D. N. Krizhanovskii, D. M. Whittaker, R. Bouchekioua, D. Sanvitto, S. Al Rizeiqi, M. S. Skolnick, P. R. Eastham, R. André, and L. S. Dang, *Phys. Rev. Lett.* (in press).
- [6] T. D. Doan, H. T. Cao, D. B. Tran Thoai, and H. Haug, *Phys. Rev. B* **74**, 115316 (2006).
- [7] R. M. Stevenson, V. N. Astratov, M. S. Skolnick, D. M. Whittaker, M. Emam-Ismail, A. I. Tartakovskii, P. G. Savvidis, J. J. Baumberg, and J. S. Roberts, *Phys. Rev. Lett.* **85**, 3680 (2000).
- [8] W. Langbein and B. Patton, *Phys. Rev. Lett.* **95**, 017403 (2005).
- [9] A. V. Andreev, V. Gurarie, and L. Radzihovsky, *Phys. Rev. Lett.* **93**, 130402 (2004).
- [10] R. A. Barankov and L. S. Levitov, *Phys. Rev. Lett.* **93**, 130403 (2004).
- [11] E. A. Yuzbashyan, O. Tsypliyatyev, and B. L. Altshuler, *Phys. Rev. Lett.* **96**, 097005 (2006).
- [12] P. R. Eastham, *J. Phys.: Condens. Matt.* **19**, 295210 (2007).
- [13] A. J. Ramsay, R. S. Kolodka, F. Bello, P. W. Fry, W. K. Ng, A. Tahraoui, H. Y. Liu, M. Hopkinson, D. M. Whittaker, A. M. Fox, et al., *Phys. Rev. B* **75**, 113302 (2007).
- [14] Y. Wu, X. Li, L. M. Duan, D. G. Steel, and D. Gammon, *Phys. Rev. Lett.* **96**, 087402 (2006).
- [15] V. S. Malinovsky and J. L. Krause, *Eur. Phys. J. D* **14**, 147 (2001).
- [16] J. Keeling, P. R. Eastham, M. H. Szymanska, and P. B. Littlewood, *Phys. Rev. B* **72**, 115320 (2005).
- [17] P. R. Eastham and P. B. Littlewood, *Phys. Rev. B* **64**, 235101 (2001).
- [18] M. H. Szymanska, J. Keeling, and P. B. Littlewood, *Phys. Rev. B* **75**, 195331 (2007).
- [19] K. Staliūnas and V. J. Sánchez-Morcillo, *Transverse Patterns in Nonlinear Optical Resonators*, no. 183 in Springer Tracts in Modern Physics (Springer, 2003).
- [20] H. Risken (North Holland, Amsterdam, 1970), vol. 8 of *Progress in Optics*, chap. 5, pp. 241 – 294.
- [21] P. R. Eastham and P. B. Littlewood, *Phys. Rev. B* **73**, 085306 (2006).
- [22] P. G. de Gennes, *Superconductivity of Metals and Alloys* (Westview, 1999).
- [23] I. A. Shelykh, M. M. Glazov, D. D. Solnyshkov, N. G. Galkin, A. V. Kavokin, and G. Malpuech, *Phys. Stat. Sol. (c)* **2**, 768 (2002).
- [24] F. P. Laussy, G. Malpuech, A. Kavokin, and P. Bigenwald, *Phys. Rev. Lett.* **93**, 016402 (2004).
- [25] H. Vinck-Posada, B. A. Rodriguez, P. S. S. Guimaraes, A. Cabo, and A. Gonzalez, *Phys. Rev. Lett.* **98**, 167405 (2007).
- [26] A. G. Steffan and R. T. Phillips, *Phys. Stat. Sol. (a)* **190**, 541 (2002).